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6 SUMMARY OF WORK ON
INITIATION, COMBUSTION AND TRANSITION
TO DETONATION IN HOMOGENEOUS AND
HETEROGENEOUS REACTIVE MIXTURES.

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ABSTRACT

This short interim report summarizes the work completed during the period June 1, 1977 to May 31, 1978 for the Air Force Office of Scientific Research as part of Grant No. AFOSR77-3336. The research deals with the broad topics of initiation, combustion and transition to detonation in homogeneous and heterogeneous reactive mixtures. One specific area deals with analytical and experimental work directed to direct initiation of detonation by a nonideal blast wave in chemically sensitized reactive fuel-air clouds. The other specific topic involves the hydrodynamic modelling of ignition and flamespreading in granular energetic solids to predict the potential for deflagration-to-detonation (DDT).

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INTRODUCTION

The direct initiation of detonation by a localized source such as a laser pulsed spark, capacitance spark, exploding wire, or exploding high explosive charge is not well understood at the present time. Initiation by the use of localized chemical accelerators is even less understood. Here, there is still the question of whether one can generate an initiating shock wave by using the proper distribution of a chemical accelerator in a source region. Also, nonlinear, two-dimensional initiation behavior is not understood. The proposed work will encompass an experimental program in which nonlinear initiation behavior will be studied in systems of direct interest to the Air Force, and a theoretical program in which the basic mechanisms of initiation by localized accelerator concentrations will be explored.

It is expected that these developments will justify the use of direct testing required to discover the initiation behavior of a wide variety of fuel-accelerator combinations. It therefore has direct applicability to Phase III FAE development as well as the studies of fuel tank vulnerability. Furthermore, the program will have application to understanding the nature of non-ideal explosions that occur during explosive release accidents.

The DDT phenomenon in granulated propellant or explosives involves a series of complex transient processes that are not well understood at the present time. It is hypothesized that the normal burning process of the solid propellant is disturbed by an abnormality such as a crack in the propellant grain. This abnormality generates regions of porous propellant which can be ignited locally, causing a pressure buildup and formation of a weak shock. If detonation is to be excited following this ignition, it is necessary to

ensure a sufficiently rapid pressure buildup. In the case of porous propellant, this may be achieved as a result of the penetration of gaseous combustion products into the interior pores of the solid, which leads to the disturbance of surface burning conditions. Thus, in this case, heat transfer by conduction is replaced by convective heat transfer. Subsequent acceleration of ignition (flame) fronts begins and pressure waves are generated which become shocks. These shocks cause large local over-pressurization and often change into a detonation.

To analyze this phenomenon, the reactive two-phase (solid, gas) conservation equations of continuity, momentum and energy must be solved along with many constitutive relations to account for heat transfer interaction, pressure losses through the aggregate, ignition criteria, unsteady burning rates, etc. It is only through solutions of such a fluid-mechanics model that one can develop criteria to specify the conditions under which the burning propellant is susceptible to transition to detonation.

SUMMARY OF PROGRESS

During this period, study continued on phenomena associated with initiation, combustion, transition to detonation (DDT) and attenuation in homogeneous and heterogeneous reactive media. A modified Lagrangian time dependent finite difference (Oppenheim (CLOUD)) program was used to study direct initiation of detonation by a nonideal blast wave in chemically sensitized reactive fuel-air clouds. Modeling of the Arrhenius kinetics throughout the explosion region in such a manner as to also satisfy computer stability requirements and reasonably short run times has been completed. Testing of different sensitizer concentration profiles in the source region has been started. The reflected shock study of initiation has been completed. The shock tube has been modified by placing a 15° ramp on a splitter plate at the back wall. Testing with this arrangement has started.

The fluid dynamics needed to predict deflagration-to-detonation transition in granulated beds of high energy solid propellant is still being modeled. We have determined the sensitivity of the inter-phase viscous drag and heat transfer on the predictions of the flamespreading rate in such packed beds. The results of this are reported as AIAA Paper 78-1013, which will be presented at the AIAA/SAE 14th Joint Propulsion Conference in July 1978. Also completed during this past year were (a) a detailed evaluation and preliminary design of experiments that would provide the necessary convective heat transfer and pressure drop relations through packed beds of solid particles, as would be needed under the high pressure/high temperature flow conditions prior to detonation transition, and (b) a new numerical integration scheme that should allow for shock-capturing analysis to be incorporated in the DDT model.

A SYNOPSIS OF WORK RELATED TO FUEL/AIR INITIATION AND BLAST WAVES

PROGRESS

In the theoretical initiation program we have adapted the Oppenheim CLOUD program, which is a constant time step Lagrangian finite difference scheme, so we can examine the question of what type of distribution of accelerator must be present in a spherical source region to generate a shock wave in the surrounding region of sufficient strength to cause direct initiation of detonation in the surrounding region. For our now completed first case, the distribution of accelerator had a uniform central core surrounded by a decreasing concentration given by

$$F(R) = F_1 [\cos(3\pi\delta) - 9.0 \cos(\pi\delta) + 8.0] / 16.0$$

where F_1 is the concentration in the central core,

$$\delta = \frac{R_0 - R}{R_0 - R_1} \quad \text{for the range } R_1 \leq R \leq R_0,$$

R_1 is the radius of the central core, and R_0 is maximum radius at which accelerator is present. In this first case we made $R_1 = 0.2 R_0$.

We first attempted to use an Arrhenius kinetic law throughout the explosion region but we found the use of this law led to rates of energy becoming so large that the numerical stability criterion required very small time steps and consequently the computation time became excessive. We then modified the law by imposing a maximum rate which could not be exceeded and in addition imposed a minimum allowable time step. After making these modifications we have been able to complete the calculations for the first case.

The total energy added before reaction would be completed in our heat addition model had a value which would produce $M_{CJ} = 7.53$. While we did not reach this value, we continued the calculation until the shock Mach number had reached a value of approximately $0.9 M_{CJ}$ and when we discontinued the calculation the Mach number was still slowly increasing. The pressure behavior shows a long incubation time of approximately 7.5 time units and the transition to $0.9 M_{CJ}$ occurred in approximately 3.0 additional time units. The most interesting graphs of time pressure behavior are given in Figs. 1a and 1b with the initial time in Fig. 1a being immediately after the incubation period had passed. The pressure rise in the uniform central core builds up very smoothly to a value which is approximately 60% of the pressure rise which would be achieved if the process had occurred at constant volume. Then in Fig. 1b, after the reaction is completed in the central core, we see the pressure begin to decay and this rarefaction propagates out to the front. But the reaction is not quenched, for the strength of the front keeps increasing. Presumably, this critical period is passed because the concentration of the accelerator is still quite high.

We caution that this successful transition to detonation may have been fortuitous because of our modified rate law. One can speculate that using a full Arrhenius law would have led to a much more rapid energy release, with a shorter time for the energy release process to have been completed, and thus more nearly approaching a constant volume process. While a higher pressure would have been achieved, in the shorter time the wave front would not have moved out so far and the rarefaction wave would overtake the front at a smaller radius. The quenching effect might then have been more severe because of the smaller radius and possibly could have stopped the reaction. To see if we can

achieve transition in another case, we are using the same rate law but have decreased the total energy to be added so that $M_{CJ} = 5.5$ approximately.

In our nonlinear initiation experimental program we have completed the numerical studies for the reflected shock initiation under nonuniform conditions. From the results of these calculations we chose a 15° ramp which now has been mounted on a splitter plate placed so that the end of the ramp is at the back wall. Testing of stoichiometric propane-air mixtures with this ramp arrangement has been started, but so far we have not succeeded in obtaining a smoke track record of the triple point trajectory of the Mach stem shock caused by the ramp or of the subsequent reflection off the back wall. We have, however, obtained side wall and end wall pressure records which indicate that detonation has occurred.

PLANS FOR THE COMING YEAR

In the theoretical program related to initiation, we expect that the strength of the shock that is generated by the region that contains the chemical accelerator will be very dependent upon both the characteristic size of the region that contains a uniform concentration of the accelerator at the center of the spherical region and the gradient of accelerator concentration in the region between the central core and the pure fuel mixture in the surrounding gas. As indicated earlier, we first have changed the total energy release so the M_{CJ} value will decrease from ~ 7.5 to ~ 5.5 which is more typical of hydrocarbon-air mixtures. We then plan to simply vary the relative size of the core and gradient region to see how the shock acceleration process is affected. We would then vary the shape of the concentration profile in the gradient region to see again how the shock acceleration process is affected. We plan to do

these calculations in stages and each parametric variation will be dependent upon what we see in the previous sequence of variations.

In the experimental program related to nonlinear initiation effects, we are using the propane-air system because of our previous experience in this system. We first are using a stoichiometric mixture and will survey the temperature range for these reflected shocks from about 900°K to about 1200°K. After making the 20 to 30 shots needed to do this, we will make a decision as to whether to try other concentrations of propane or whether to try other fuels. Since we are primarily concerned with reactive exothermic gas dynamic processes, this study will not simply be a routine testing technique for different types of fuels.

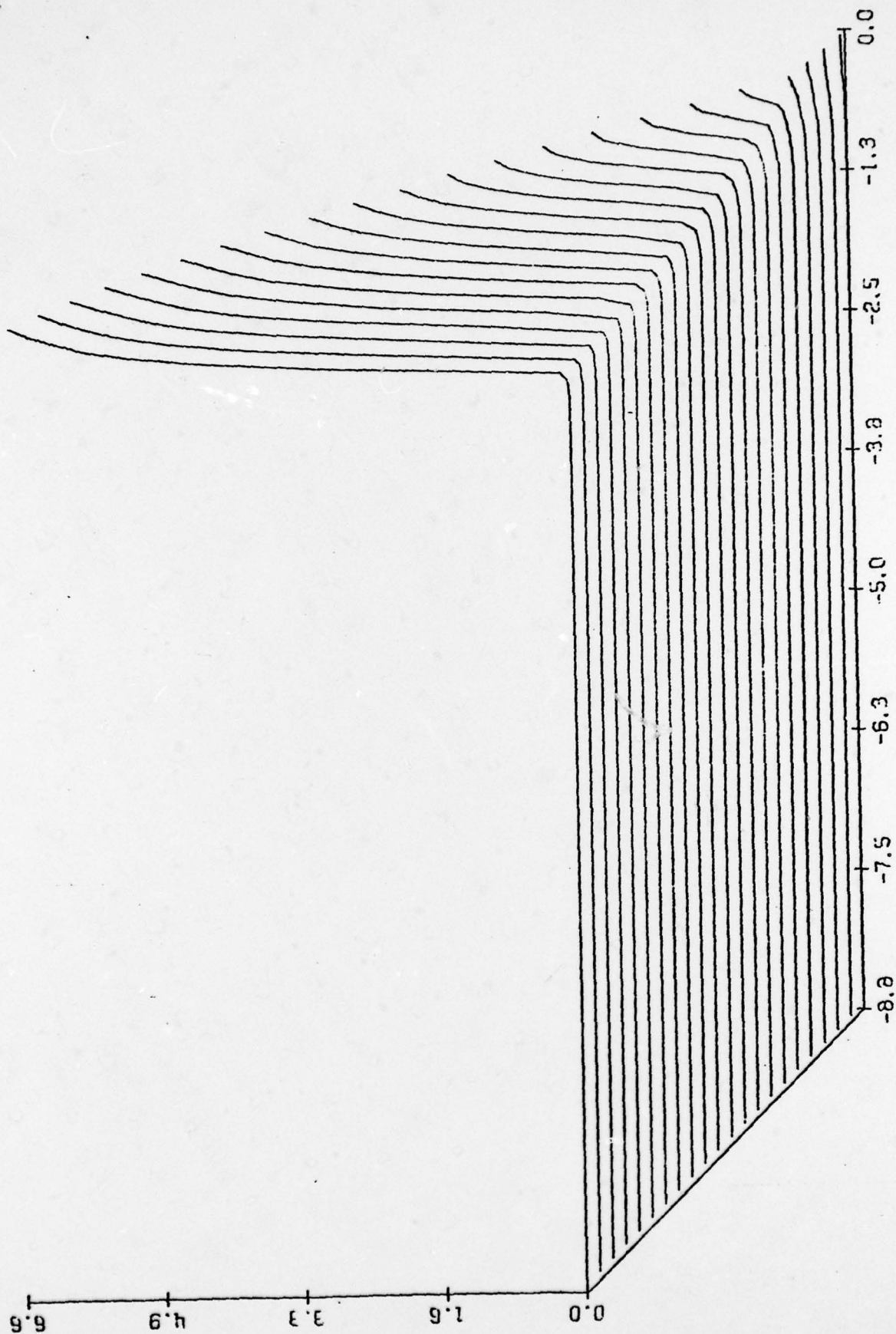


FIG. 1A PRESSURE / PO DISTRIBUTION VS. DISTANCE / DO AND TIME / TO

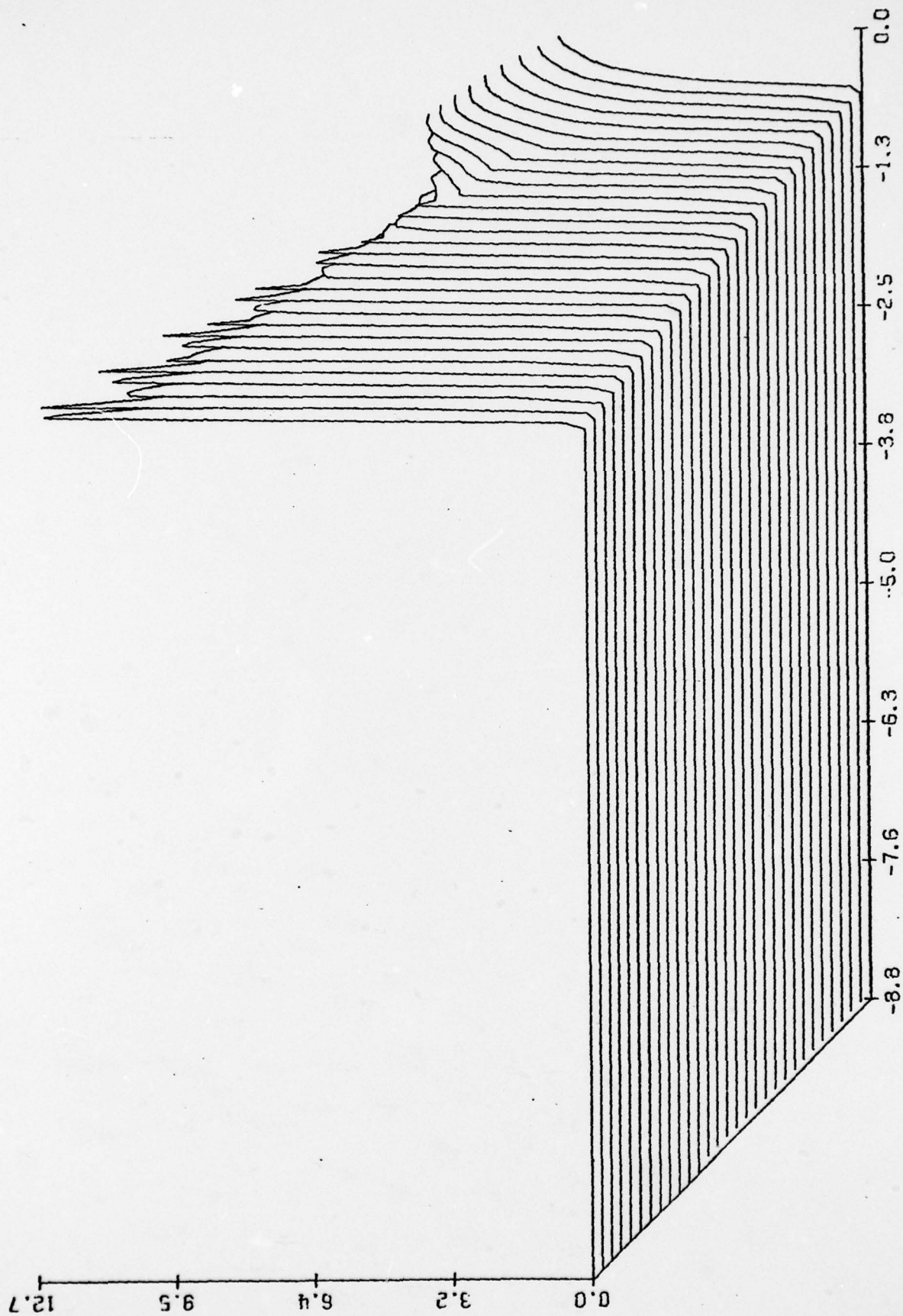


FIG. 1B PRESSURE / PO DISTRIBUTION VS. DISTANCE / DO AND TIME / TO

A SYNOPSIS OF MATHEMATICAL MODELING
TO STUDY DDT IN GRANULATED ENERGETIC SOLIDS*

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ABSTRACT

This short paper will review and highlight the work reported in the open-literature on convective mode combustion modeling through granulated energetic solids which have been used to study the potential for deflagration-to-detonation transition (DDT). Typical results are presented which clearly indicate that the convective process is a rapidly accelerating one, producing high internal gas pressures with shock-like features. Specific references will be sighted with emphasis on the limits inherent in the theoretical modeling work. The paper concludes with recommendations for basic research that is needed, before this type of combustion-fluid mechanics analysis can be expected to provide quantitative predictions for DDT potential.

INTRODUCTION

The transition from combustion to detonation in condensed, but porous propellants and explosives has in recent years received attention both from analytical and experimental efforts. A large amount of the experimental work has been carried out by Soviet scientists, much of which has been documented by A. F. Belyaev et al. [1] and recently reviewed by H. H. Bradley and T. L. Boggs [2]. The work of R. R. Bernecker and D. Price [3,4] represents detailed and specific experimental research toward understanding deflagration-to-detonation transition (DDT) in porous explosives. These latter studies clearly indicate that the buildup to detonation represents a coupling between the pressure (shock or compression) fronts and the convectively driven flame front.

As would be expected if detonation is to follow ignition (and deflagration) it is necessary, above all, to ensure a sufficiently rapid pressure buildup. In a porous combustible system, this is achieved as a result of the penetration of the gaseous combustion products into the porous interior which leads to accelerating ignition of additional burning surfaces. Thus heat transfer by conduction is replaced by convective heat transfer. The specific mechanism to achieve DDT is yet to be fully understood, but proposed phenomenological mechanisms have been proposed by Bernecker and Price [3], by R. W. Van Dolah et al. [5], and by Belyaev et al. [1].

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Prior to the modeling work used to analyse the DDT process, a considerable amount of two-phase convective combustion modeling had been performed by Kuo and Summerfield [6,7], Kuo, et al [8], Gough [9], Van Tassell and Krier [10], and Krier, et al [11] applied to ignition and combustion in packed beds of gun-propellant grains. Although much of the formalism for such types of unsteady, two-phase, reactive flows is similar to the flow in the DDT problem, significant differences do exist. These would include the fact that, (a) the time scale in the DDT problem for the accelerating convectively driven front is several orders of magnitude less than the millisecond scale events encountered in the pressure wave and ignition transient in gun cartridges, and (b) the gas pressures are greater (by at least one order of magnitude) because of the more energetic solids considered and their smaller granulation.

The first real application of the general logic used in previous two-phase flow models, ie., Ref. 6-11, was attempted by Beckstead, et al [12]. That work was built upon the code used by Krier and co-workers at the University of Illinois, but with many important modifications in both the formulation of the governing conservation equations (as had been used in Refs. 10 and 11) and in the initial conditions modeled. One of the important aspects presented in the work by Beckstead, et al [12] was the development of a critical condition that defines the runup length to detonation. There, use was made of the concept of a "critical energy" for initiation to detonation, as originally proposed by Walker and Wasley [13].

Additional DDT calculations were carried at Hercules, Inc. by Pilcher, et al [14], using a fluid mechanics model similar to that described in Ref. 12, but with the additional feature of linking the motion of the confining walls of the bed to the transient internal pressure. A comparison of the model predictions to detonation, with tests in a variety of configurations using granulated, class A HMX showed very good agreement.

Alternate DDT models were developed at the University of Illinois by Krier and co-workers [15-17] which clearly indicated that the transient two-phase flow in confined granulated propellants could lead to accelerating deflagration fronts that approached detonation speeds. Table 1 presents the list of references of these analytical models, as they were applied to study the DDT process. Details of the assumptions made in those models are too numerous to repeat here. However, the following section enumerates those aspects where this author believes that the analyses must be improved. Basically these improvements can be summarized by the statement that (a) there is very little high pressure two-phase flow work that is directly applicable to the transient processes one needs to model transition to detonation, and (b) the ignition and burning rate data must be extrapolated from conditions no-where near that needed in the DDT process.

TABLE I

Listing of DDT Convective Burning Models

1. Beckstead, M. W., Peterson, N. L. Pilcher, D. T., Hopkins, B. D., and Krier, H., "Convective Combustion Modeling Applied to Deflagration-to-Detonation Transition of HMX," Combustion and Flame, Vol. 30, pp. 231-241 (1977).
2. Pilcher, D. T., Beckstead, M. W., Christensen, L. W., and King, A. J., "A Comparison of Model Predictions and Experimental Results of DDT Tests," AIAA Paper 77-856, presented at the 13th AIAA/SAE Joint Propulsion Conference, Orlando, FL (July 1977).
3. Krier, H. and Gokhale, S. S., "Modeling of Convective Mode Combustion through Granulated Propellant to Predict Detonation Transition," AIAA Journal, Vol. 16, pp. 177-183 (February 1978).
4. Krier, H., Gokhale, S. S., Hoffman, S. J., "Unsteady Two-Phase Flow Analysis Applied to Deflagration-to-Detonation Transition," AIAA Paper 78-1013, presented at the 14th AIAA/SAE Joint Propulsion Conference, Las Vegas, NV (July 1978).

WHERE MODELS NEED IMPROVEMENT

When reviewing the papers listed in Table 1, it is immediately obvious where one can begin to question the general applicability of the analysis. Assuming that the quasi, one-dimensional formulation of the continuity, momentum, and energy equations as presented in those models are generally correct, the problems arise in the assumed constitutive relations inputted into the models. (The reason for the differences in the expression of the two-phase conservation equations are discussed in References 15 and 17.)

The key constitutive relations, in some representative order of their importance (as they effect the predictions) are:

- (1) Gas-particle viscous interaction, which basically determines the hot-gas permeability into the unignited portions of the granulated bed.
- (2) Gas-particle heat transfer coefficient, which determines the convective energy transfer from the turbulent flow hot gases to particles.
- (3) The propellant burning rate at extreme pressures (of the order of 10^9 nt/m²) and rapid rates of pressure change, dp/dt.
- (4) The ignition criteria that fixes the time that the particles are ignited by the convective processes. Heat flux rates often exceed 10^9 watts/m².
- (5) The intergranular stress that, under highly transient conditions, limits the (extreme) particle compaction.

- (6) The gas equation-of state. It has been determined that a constant co-volume correction to the ideal e.o.s. is not valid at these high pressures.
- (7) The temperature dependency on the specific heat, gas viscosity, and gas conductivity.
- (8) A DDT criterion

Recently the study reported in Ref. 17 provides evidence that the gas-particle drag interaction, as generally used in the DDT models, can lead to sizeable particle motion and extreme compaction, unless one can provide the appropriate intergranular resistance to the compaction. In short, it was not possible to properly predict the convective mode combustion dynamics in long granulated beds, using the same gas-particle drag interaction-law that seemed to work for the shorter beds. And as discussed in Ref. 17, (and elsewhere), the data base that has been used to correlate the pressure-drop in packed beds has been carried out both at steady-state conditions and at Reynold's numbers several orders of magnitude less than that needed for the DDT flow processes. In addition, these correlations were based on tests only with inert particles.

The same criticism must obviously be applied to the heat transfer coefficient, as correlated at low pressures and relatively low velocities, resulting in only moderate Reynold's number ranges. From the sensitivity studies reported in Refs. 15-17, a general conclusion can be stated that both the gas-particle friction coefficient and heat transfer coefficient, when extrapolated to the Reynold's number and low porosities needed in the DDT flows, are too large, by at least one order of magnitude. Propellant and explosive linear burning rates are generally expressed as functions of the ambient pressure, through a power law steady-state correlation. Measurement of burning rates are generally never carried out at pressures greater than 10,000 - 20,000 psi. Yet the pressures predicted to occur in the confined granulated bed during the flamespreading can exceed 250,000 psi (1.72×10^9 nt/m²). And the pressure can change with time at rates of the order of 1000 psi/ μ sec to 10,000 psi/ μ sec! Thus it is questionable whether steady state burning rates, extrapolated from much lower pressures represent the dynamic burning rates during these flamespreading processes. Also the gas velocities relative to the particles can vary from 10 to 1000 meters/second, bringing unknown factors, such as erosive burning augmentation into the burning rate expression.

Moving down the list of the eight key constitutive relations used in the DDT models, one also requires an ignition criterion from the stimulus of the convective heat transfer from the hot gaseous combustion products as they are forced through the unignited regions. It was already mentioned that heat fluxes ranging from 100 to 1000 BTU/in² sec are calculated to occur during the DDT process. Little, if any, data exists at these extreme flux rates that ties into either a critical propellant surface temperature or a critical ignition energy. With these flux rates, ignition delay times are often less than several microseconds. No data exists to verify such a possibility.

Finally it is assumed that one has a good handle on the high pressure (high temperature) equation of state as well as the several gas transport parameters listed in item #7.

14

SOME TYPICAL RESULTS

Figures 1-3 present the pressure wave history, the temperature front histories, and the ignition (flame) front locus, respectively for a transient flamespreading event of relatively high energy, but small particle granulated bed. The specific details of that calculation are given in Ref. 17; the bed was initially packed with 60% solids loading ($\phi_0 = 0.40$). These results are representative of those presented in the other DDT models (listed in Table 1). Flame fronts are predicted to accelerate to speeds of 2mm/ μ sec within 5-6 cm, and pressures can exceed several G nt/m². Generally it is still not possible to predict to abrupt transition in flame fronts (speeds ranging from 6-8 mm/ μ sec) as observed in the DDT experiments of Bernecker and Price [3,4]. It should be noted that the experiments discussed in Refs. 3 and 4 were carried out with fairly long granulated beds (25 cm) with solids loadings of 70 to 90 percent. Such high solids fraction require multi-modal particle size distribution. The DDT models to date have considered only one representative particle size, forcing initial solids loadings to be no greater than 75%.

CONCLUDING REMARKS

It is fair to state that for such a complex highly transient flow process, the models that have been developed are limited in their description of the flow physics. The reasons for this has been briefly outlined above. Improvements at this point will require basic two-phase flow experiments at conditions of interest to the DDT regime, a sizeable investment in time and money. In addition propellant ignition and burning rate experiments must be devised for the appropriate energy fluxes and pressure ranges.

However, it is also fair to conclude that the models in their present state have already been useful in indicating the potential for detonation for certain high energy, rapid burning rate propellants packed at significant granulation. The best example to date of the usefulness that such models bring is obvious in the results reported by Pilcher, et al [14]. There is no need to add that a significant amount of work is still required to use the mathematical models for accurate quantitative predictions for transition to detonation.

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15

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17. Krier, H., Gokhale, S. A. and Hoffman, S. J., AIAA paper 78-1013, presented at the 14th AIAA/SAE Joint Propulsion Conference, Las Vegas, NV, July 1978.

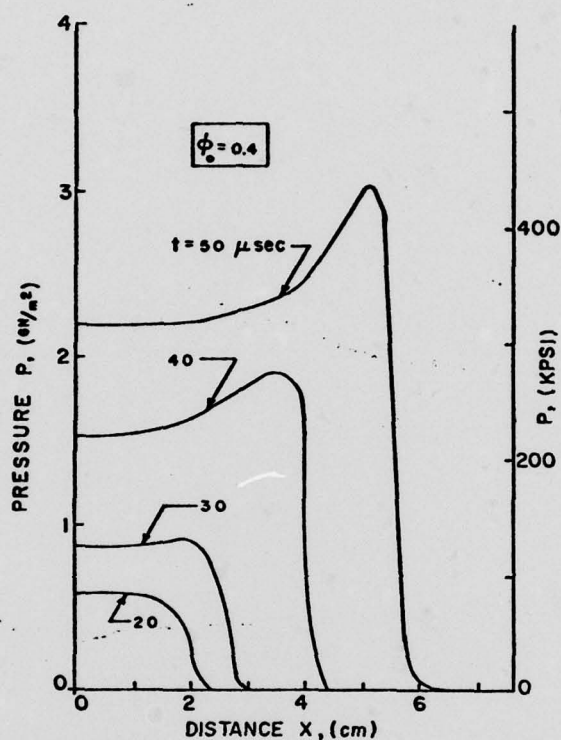


Figure 1. Pressure distribution during the accelerating deflagration in a bed initially packed at 60% solid loading with 200 μm diameter energetic propellant grains. Propellant heat of combustion is 1350 cal/gm and the assumed burning rate (cm/sec) is $4 (10^{-7}) P^{0.9}$ where $[P] = [\text{nt/m}^2]$.

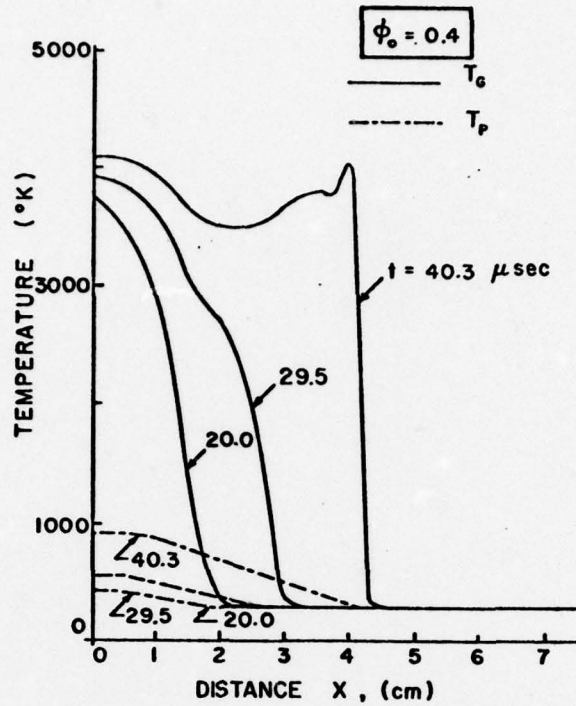


Figure 2. The gas and particle temperature distribution development for the pressure history shown in Fig. 1.

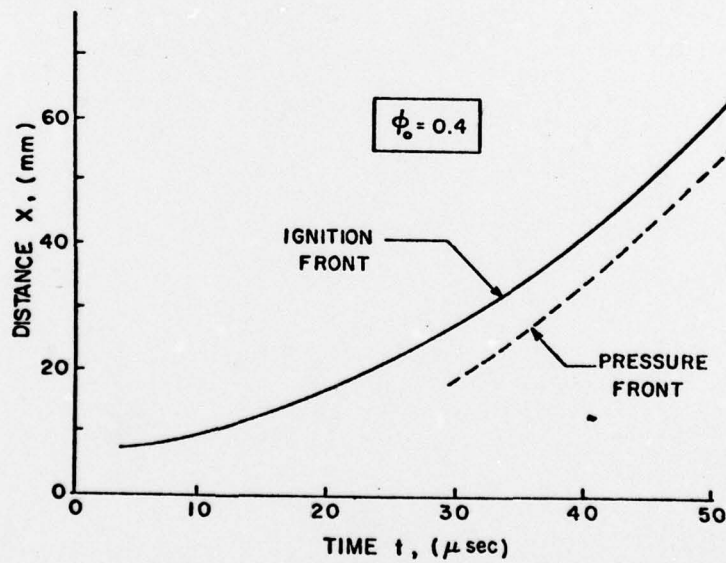


Figure 3. Flame front locus (with the pressure front) for the pressure and temperature history shown in the previous two figures.

SUMMARY OF PUBLICATIONS/REPORTS

1. "The Effect of Additives on the Ignition Delay Time for Stoichiometric Propane-air Mixtures," by Gregory M. Maegher, M.S. Thesis, Department of Aeronautical and Astronautical Engineering, University of Illinois, (under the direction of R. A. Strehlow) (1978).
2. "Direct Initiation of Detonation," by R. A. Strehlow and H. O. Barthel, University of Illinois Report, UILU-Eng 77 0515 (1977), AFOSR TR-77-1272.
3. "Two-Phase Reactive Particle Flow Through Normal Shock Waves," by H. Krier and A. Mozaffarian, Int'l. J. Multiphase Flow, Vol. 4, pp. 65-79 (1978).
4. "Modeling of Convective Mode Combustion Through Granulated Propellant to Predict Detonation Transition," by H. Krier and S. S. Gokhale, AIAA J., Vol. 16, No. 2, pp. 177-183 (1978).
5. "Unsteady Two-Phase Flow Analysis Through Reactive Particles to Predict DDT," by H. Krier, S. S. Gokhale, and S. J. Hoffman, AIAA Paper 78-1013. Presented at the AIAA/SAE-14th Joint Propulsion Conference, Las Vegas, NV (July 1978).
6. A Final Report on DDT for work done under AFOSR 77-3115, by Krier, H. and Kezerle, J. A., was submitted and 80 copies distributed (Tech Report AFOSR-TR-78-007).

TALKS/PRESENTATIONS ON AFOSR SPONSORED WORK

A. By Roger A. Strehlow

1. Blast Waves Generated from Constant Velocity Flames, 6th Gasdynamics Symposium, Stockholm, August 1977.
2. Gaseous Detonations, DOD Workshop entitled "New Opportunities for Research in Energetic Materials," Wrightsville Beach, NC, November 1977.
3. Unconfined Vapor Cloud Explosion, Seminar at the University of Leeds, Leeds, England, August 1977.
4. Seminar on Unconfined Vapor Cloud Explosions given at the Shell Thornton Research Centre, Chester, England, August 1977.
5. Seminar entitled "Accidental Explosions," presented in London, England, to a group of insurance and technical people, August 1977.
6. Seminar entitled "Unconfined Vapor Cloud Explosions," presented to TNO in Holland, The Netherlands, August 1977.
7. "Initiation of Detonation in Fuel-Air Mixtures by Non-Linear Finite Amplitude Waves and Chemical Reactive Promoters," AFOSR Contractors' Meeting, November 1977, Champaign, IL.
8. Seminar entitled "Unconfined Vapor Cloud Explosions," presented to the Lawrence Livermore Laboratories, Livermore, CA, January 1978.
9. Seminar entitled "Detonation Structure and Initiation," given to Lawrence Livermore Laboratories, January 1978.

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) This short interim report summarizes the work completed during the period 1 June 1977 to 31 May 1978 for the Air force Office of Scientific Research as part of Grant No. AFOSR-77-3336. The research deals with the broad topics of initiation, combustion and transition to detonation in homogeneous and heterogeneous reactive mixtures. One specific area deals with analytical and experimental work directed to direct initiation of detonation by a nonideal blast wave in chemically sensitized reactive fuel-air clouds. The other specific topic involves the hydrodynamic modelling of ignition and flamespreading in granular energetic solids to predict the potential for deflagration-to-detonation (DDT).		

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